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Cooper-pair propagation and superconducting correlations in graphene

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We investigate the Cooper-pair propagation and the proximity effect in graphene under conditions in which the distance L between superconducting electrodes is much larger than the width W of the contacts. In the case of undoped graphene, supercurrents may exist with a spatial decay proportional to W^2/L^3. This changes upon doping into a $1/L^2$ behavior, opening the possibility to observe a supercurrent over length scales above 1 μm at suitable doping levels. We also show that there is in general a crossover temperature $T^* \sim v_F/k_B L$ that marks the onset of the strong decay of the supercurrent, and that corresponds to the scale below which the Cooper pairs are not disrupted by thermal effects during their propagation.

The recent interest in the physics of graphene has arisen from the observation of a number of novel electronic properties[1, 2], which are the consequence of the relativistic-like character of the electron quasiparticles. This is certainly the case of the anomalous quantization of the Hall conductivity[3, 4, 5], as well as of the existence of a finite lower bound in the conductivity at the charge neutrality point[4, 6, 7, 8]. The graphene system is unique in that the low-energy excitations have conical dispersion around discrete Fermi points, being therefore governed by a Dirac equation for massless chiral particles. The appearance of an additional pseudo-spin quantum number intrinsic to the Dirac spectrum has led to propose other unconventional effects, like the selective transmission of electrons through *n*-*p* junctions[9] or a peculiar form of Andreev reflection at the metal-superconductor interface in graphene[10].

Recently the proximity effect has been investigated in graphene in two different experiments[11, 12]. In both of them a 2D carbon layer has been contacted with superconducting electrodes, though the size and aspect ratio of the respective samples have been quite different in the two cases. Thus, while the separation of the electrodes in the experiment of Ref. 11 appears to be of the order of ~ 0.5 μm, the distance between them in the report of Ref. 12 seems to have a minimum value of about 2.5 μm. This may explain why a supercurrent has been measured between the electrodes in the first case, while the most relevant observation in the second experiment has been an abrupt drop in the resistance, below the critical temperature of the superconducting electrodes.

This crucial dependence on the distance between the electrodes may be however somewhat surprising, taking into account that supercurrents have been measured in carbon nanotubes placed between superconducting contacts, with nanotube lengths as large as ≈ 1.7 μm [13]. It becomes then pertinent to analyze the proximity effect in graphene, specially for large separation between the superconducting electrodes, in order to find out the features related to the 2D character of the material. The Josephson effect has been studied before in graphene strips with length L small relative to their width and the superconducting coherence length[14]. Here we will face the opposite situation, in which the relevant signatures are dictated by the propagation of the Cooper pairs in graphene and its dependence on thermal and interaction effects.

We investigate then the proximity effect in graphene, under conditions in which the distance L between the superconducting electrodes producing the Cooper pairs is much larger than the width W of the contacts. More specifically, we will consider that the hamiltonian for graphene with superconducting contacts along the coordinates $x_1 = 0$ and $x_2 = L$ is given by

$$\begin{aligned} H &= v_F \int d^2r \; \Psi_\sigma^{(a)\dagger}(\mathbf{r}) \boldsymbol{\sigma}^{(a)} \cdot \boldsymbol{\partial} \, \Psi_\sigma^{(a)}(\mathbf{r}) \\ &\quad + \sum_{j=1,2} t \int_0^W dy \; \Psi_\sigma^{(a)\dagger}(x_j, y) \Psi_{Sj,\sigma}(x_j, y) + \text{h.c.} \end{aligned} \quad (1)$$

$\boldsymbol{\sigma}^{(a)}$ being different sets of Pauli matrices for $a = 1, 2$ [3]. In the above expression, a sum is implicit over the spin index σ, as well as over the index a accounting for the two different valleys and corresponding Dirac spinors $\Psi^{(a)}$ at opposite K points in graphene. We will consider that the coupling to the electron fields Ψ_{S1} and Ψ_{S2} in the superconducting electrodes takes place along a segment of width W in each case. For our purposes, it will be enough to describe the superconductors in terms of the order parameter Δ and the normal density of states N.

We are going to deal in particular with the case in which the time of propagation of the Cooper pairs between the contacts is much larger than $1/|\Delta|$. This implies, equivalently, that the distance L has to be much larger than the superconducting coherence length ξ. Then, a supercurrent may arise mainly from processes in which the Cooper pairs tunnel from one of the superconductors to the 2D layer, propagating to the other superconducting contact. The Josephson current I_s can be computed as the derivative of the free energy $\mathcal{F}$ with respect to the difference χ between the respective phases

χ_1 and χ_2 of the order parameters in the two superconductors, $I_s = -2e\partial\mathcal{F}/\partial\chi$. Under the assumption of a relative large $|\Delta|$, we can make the approximation

$$\langle\Psi_{Sj,\sigma}(x_j,y;-i\tau_1)\Psi_{Sj,-\sigma}(x_j,y;-i\tau_2)\rangle \approx e^{i\chi_j}N\delta(\tau_1-\tau_2) \tag{2}$$

where the statistical averages, at temperature T, are taken over ordered products with respect to imaginary time τ. Then, the maximum supercurrent (critical current) becomes to lowest order in the tunneling amplitude

$$\begin{aligned} I_c(T) \;\approx\; & 2eN^2t^4W^2\int_0^W dy_1\int_0^W dy_2\int_0^{1/k_BT}d\tau \\ & \langle\Psi_\uparrow^{(a)\dagger}(0,y_1;0)\Psi_\downarrow^{(-a)\dagger}(0,y_1;0) \\ & \times\Psi_\uparrow^{(a)}(L,y_2;-i\tau)\Psi_\downarrow^{(-a)}(L,y_2;-i\tau)\rangle \end{aligned} \tag{3}$$

In deriving Eq. (3) we have already assumed that $L \gg W$, averaging over the separation of the electrons in the pair tunneling along the superconducting contact.

The determination of the critical current is therefore reduced to the evaluation of the propagator of the Cooper pairs in the graphene layer. The Fourier transform of this object, that we will denote by $D(\mathbf{k},i\overline{\omega})$, can be obtained from the standard diagrammatics for Dirac fermions. Here we stress that, for the sake of preserving the relativistic-like invariance, it is convenient to regularize the diagrams at high energies by using a method that maintains the space-time symmetry of the theory, like the analytic continuation in the number of dimensions[15]. In particular, the Cooper-pair propagator can be obtained in the noninteracting theory from the convolution of two Dirac propagators, and at $T=0$ it turns out to be

$$D^{(0)}(\mathbf{k},i\overline{\omega})\Big|_{T=0} = -\frac{1}{8v_F^2}\sqrt{v_F^2\mathbf{k}^2+\overline{\omega}^2} \tag{4}$$

The temperature dependence of this object can be obtained using the Matsubara formalism. In the static limit we get

$$\begin{aligned} D^{(0)}(\mathbf{k},0) \;=\; & -\frac{1}{2\pi v_F}|\mathbf{k}|\int_0^1 dx\sqrt{x(1-x)} \\ & \times\tanh(v_F|\mathbf{k}|\sqrt{x(1-x)}/2k_BT) \\ & -\frac{1}{\pi v_F^2}k_BT\int_0^1 dx \\ & \times\ln[2\cosh(v_F|\mathbf{k}|\sqrt{x(1-x)}/2k_BT)] \end{aligned} \tag{5}$$

The knowledge of the Cooper-pair propagator at zero frequency is enough to compute the critical current in Eq. (3). Interaction effects can be incorporated by summing up the different perturbative orders obtained by iteration of the scattering of the particles in the pair. If we take an average of the potential V between the particles, we have that the Cooper-pair propagator can be represented in the T-matrix approximation by

$$D(\mathbf{k},\omega) \approx \frac{D^{(0)}(\mathbf{k},\omega)}{1+VD^{(0)}(\mathbf{k},\omega)} \tag{6}$$

We remark anyhow that the interaction effects cannot affect significantly the propagation of the Cooper pairs at low temperatures. When there is exchange of the valleys in the scattering of the electrons in the pair, the Coulomb potential V_C becomes suppressed by a large momentum-transfer $2k_F$, down to $V_C \sim e^2/2k_F$. At the small values of $|\mathbf{k}|$ relevant for the long-distance regime, the denominator in Eq. (6) remains then very close to 1. In the opposite case of no valley exchange, we observe that at low momentum-transfer $V_C D^{(0)}$ has a relative strength of the order of $\sim e^2/v_F$. At low energies, this effective coupling is strongly renormalized, down to a value which may be about one order of magnitude below the nominal coupling[16]. We have checked that, in practice, this makes very small the difference between computing the critical current (3) from either $D(\mathbf{k},0)$ or $D^{(0)}(\mathbf{k},0)$, at the relevant temperatures in the experiments (of the order of ~ 1 K).

It becomes convenient to factor out from (3) the relative tunnel conductances of the interfaces, which are given each by the constant quantity Nt^2W/v_F [17]. We will deal then with the intrinsic 2D dependence of the critical current, which becomes in terms of the Fourier transform of the Cooper-pair propagator

$$\begin{aligned} I_c^{(2D)}(T) \;\approx\; & 2eW^2v_F^2 \\ & \times\int_0^\infty\frac{dk}{2\pi}\,|\mathbf{k}|\;J_0(|\mathbf{k}|L)D(\mathbf{k},0)e^{-|\mathbf{k}|/k_c} \end{aligned} \tag{7}$$

where k_c is a short-distance cutoff, that we take of the order of $\sim 1\ \mathrm{nm}^{-1}$.

The critical current computed from (7) displays a number of peculiar features. From the scaling of $D^{(0)}(\mathbf{k},0)$ in the zero-temperature limit, it is easily seen that $I_c^{(2D)}(0)$ must decay as $\sim v_FW^2/L^3$ at large separation between the contacts. Another important property is that there is always a crossover temperature $T^* \sim v_F/k_BL$ which marks the onset of a power-law decay of the supercurrent at high temperatures. This is shown in Fig. 1, which represents the behavior of the critical current as a function of T for $L = 5\times10^3/k_c$ and $W = 10^2/k_c$. The dependence of $I_c^{(2D)}(T)$ is qualitatively similar to what is found in carbon nanotubes with long separation between superconducting contacts[18, 19]. The crossover temperature corresponds to the scale at which the Cooper pairs fail to propagate efficiently between the electrodes, as they become disrupted along the way by thermal effects.

The novel behavior of the supercurrents in graphene with respect to that in the carbon nanotubes comes from the different scaling with length L at low temperatures. This is given by a $1/L^3$ power-law behavior in graphene, instead of the $1/L$ scaling of a noninteracting 1D system[18]. We may consider for instance the sample described in Ref. 12, with an approximate separation between the electrodes $L \approx 2.5\ \mu$m. The plot in Fig. 1 gives the theoretical values of the critical current,

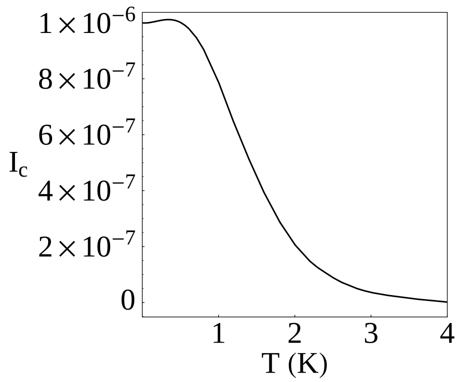

FIG. 1: Plot of the critical current $I_c^{(2D)}(T)$ (in units of $10^{-2}ev_Fk_c \approx 1.2$ μA) for $L = 5 \times 10^3/k_c$ (≈ 2.5 μm) and $W = 10^2/k_c$ (≈ 50 nm).

in units of $10^{-2}ev_Fk_c \approx 1.2$ μA. We observe that, in the low-temperature regime to the left of the crossover, the expected critical currents should have a magnitude $I_c^{(2D)} \sim 10^{-3}$ nA. Such an small scale may explain the difficulty to establish a supercurrent in graphene when there is a separation of the order of microns between the superconducting electrodes.

On the other hand, the crossover shown in Fig. 1 has a remarkable correspondence with the abrupt drop in the resistance reported in Ref. 12. This feature has been observed at a temperature of about 1 K, and it does not seem to bear a direct relation to the critical temperature T_c (≈ 4 K) of the superconducting electrodes. We observe that the crossover displayed in Fig. 1 corresponds to a temperature of about 1.5 K, in fair agreement with the position of the drop measured experimentally in the resistance. It is therefore quite likely that this feature may be an indirect signature of the temperature up to which the Cooper pairs are able to propagate without disruption, along the 2.5 μm-long path of the sample reported in Ref. 12.

The fast decay of the supercurrent computed from (7) can be traced back to the vanishing density of states of graphene at the charge neutrality point. Actually, the form of the propagator in (4) is a direct consequence of the conical dispersion around the Fermi points of graphene. One can therefore expect important changes in the Cooper-pair propagation upon doping the electron system. This can be investigated formally by introducing a finite chemical potential μ, with the aim of shifting the Fermi level away from the charge neutrality point. At $\mu > 0$, the Dirac propagator can be written as[20]

$$G^{(0)}(\mathbf{k},\omega) = (\omega + v_F\boldsymbol{\sigma}\cdot\mathbf{k})[\frac{-1}{-\omega^2 + v_F^2\mathbf{k}^2 - i\epsilon} + i\pi\frac{\delta(\omega - v_F|\mathbf{k}|)}{v_F|\mathbf{k}|}\theta(\mu - v_F|\mathbf{k}|)] \quad (8)$$

which is a convenient alternative form of expressing the electron propagator

$$G^{(0)}(\mathbf{k},\omega) = \frac{\theta(\varepsilon(\mathbf{k}) - \mu)}{\omega - \varepsilon(\mathbf{k}) + i\epsilon} + \frac{\theta(\mu - \varepsilon(\mathbf{k}))}{\omega - \varepsilon(\mathbf{k}) - i\epsilon} \quad (9)$$

for a dispersion with two branches $\varepsilon(\mathbf{k}) = \pm v_F|\mathbf{k}|$. The Cooper-pair propagator can be computed now from the convolution of two Dirac propagators like (8). The result at $\omega = 0$ is

$$\begin{aligned} D^{(0)}(\mathbf{k},0) &= -\frac{1}{2\pi v_F^2}\mu \qquad \text{if} \quad |\mathbf{k}| < 2\mu \\ &= -\frac{1}{8v_F}|\mathbf{k}| + \frac{1}{4\pi v_F}|\mathbf{k}| \arcsin\left(\frac{2\mu}{|\mathbf{k}|}\right) \\ &\quad -\frac{1}{2\pi v_F^2}\mu \quad \text{if} \quad |\mathbf{k}| > 2\mu \end{aligned} \quad (10)$$

The slight change produced by the chemical potential in the infrared behavior of $D^{(0)}(\mathbf{k},0)$ is enough to modify the long-distance decay of the supercurrent. We have represented in Fig. 2 the result of evaluating $I_c^{(2D)}(0)$ from Eq. (7) with the Cooper-pair propagator in (10). It can be seen that, at $L \sim v_F/\mu$, the power-law decay of the critical current changes from $1/L^3$ to $1/L^2$. The supercurrents cannot be enhanced anyhow up to the magnitudes that they reach in a 1D electron system, where the decay is given by a $1/L$ dependence in the noninteracting theory. However, it may be worthwhile to explore experimentally the consequences of doping the graphene layer, up to levels where it can be affordable to measure supercurrents of the order of ~ 1 nA for suitably small values of L, according to the plot shown in Fig. 2.

We also remark that doping the graphene layer may be the way to obtain experimental signatures of dynamical superconducting correlations. The pertinent approach to address this question is to deal again with the theory at nonvanishing chemical potential, starting from the Dirac propagator in Eq. (8). Computing now the Cooper-pair propagator at real frequency ω, the possible superconducting instabilities have to reflect as singularities in $D(0,\omega)$, as it happens with the formation of bound electron pairs in the BCS theory[21].

The evaluation of the Cooper-pair propagator at chemical potential $\mu > 0$ gives the result

$$\begin{aligned} D^{(0)}(0,\omega) &= -\frac{1}{4\pi v_F^2}\omega \log\left|\frac{2\mu - \omega}{\omega}\right| \\ &\quad -\frac{1}{2\pi v_F^2}\mu - i\frac{1}{8v_F^2}\omega \end{aligned} \quad (11)$$

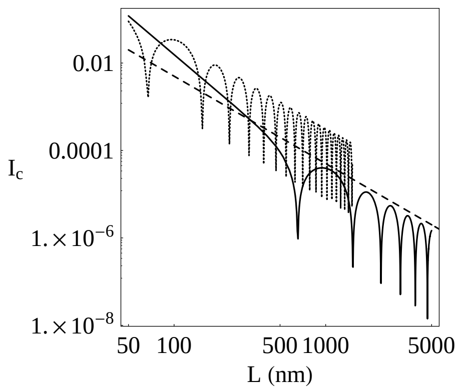

FIG. 2: Plot of the zero-temperature critical current $I_c^{(2D)}(0)$ (in units of $10^{-2}ev_Fk_c \approx 1.2\ \mu$A) as a function of the distance L (keeping $W = 10^2/k_c \approx 50$ nm), for chemical potential $\mu = 1$ meV (full line) and $\mu = 0.01$ eV (dotted line). The dashed line corresponds to a $1/L^2$ decay and it is drawn here as a reference of that power-law behavior.

We may sum up again multiple scattering processes of the Cooper pairs in the framework of the T-matrix approximation. We observe then that $D(0,\omega)$ has a pole under conditions where the dominant interaction is attractive, such that $V < 0$. The denominator $1 + VD^{(0)}(0,\omega)$ in (6) vanishes at values of the frequency ω given by the equation

$$|2\mu - \omega| = \omega \exp\left(\frac{4\pi v_F^2 - 2\mu V}{\omega V}\right) \tag{12}$$

There is always a solution of Eq. (12) for ω below and close to 2μ when $V < 0$. Such a frequency marks the formation of bound electron pairs, that may take place for arbitrarily small values of μ, despite the fact that the density of states of graphene vanishes at $\omega = 0$. The main difference with a conventional Fermi liquid is that the energy scale in front of the exponential in Eq. (12) may be rather small, of the order of $\sim 2\mu$.

In conclusion, we have studied the proximity effect in graphene when the distance between the superconducting contacts is much larger than their width. We have seen that, in the case of undoped graphene, the supercurrents have a fast spatial decay, proportional to W^2/L^3. This strong dependence on L reminds of the behavior of the critical current in long diffusive junctions[22]. On the other hand, it is unlikely that the experimental measures may be strongly affected by disorder in a graphene layer, since backscattering arises only from small-range scatterers with a size not larger than the lattice constant[23]. We have actually seen that the results reported in Ref. 12 are consistent with a crossover to the strong decay of the critical current at a temperature $T^* \sim v_F/k_BL$, and not at the much smaller scale given in a diffusive junction in terms of the diffusion constant $\mathcal{D}$ by $T^* \sim \mathcal{D}/k_BL^2$.

We have seen that shifting the Fermi level away from the charge neutrality point changes the dependence of the critical current into a $1/L^2$ behavior, opening the possibility to observe a supercurrent over length scales above 1 μm at suitable doping levels. We have shown that this becomes feasible anyhow below the crossover temperature T^* marking the onset of the power-law decay of the supercurrent, and that corresponds to the scale above which the Cooper pairs are increasingly disrupted by thermal effects during their propagation.

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[1] K. S. Novoselov, A. K. Geim, S. V. Morozov, D. Jiang, M. I. Katsnelson, I. V. Grigorieva, S. V. Dubonos and A. A. Firsov, Nature **438**, 197 (2005).
[2] Y. Zhang, Y.-W. Tan, H. L. Stormer and P. Kim, Nature **438**, 201 (2005).
[3] Y. Zheng and T. Ando, Phys. Rev. B **65**, 245420 (2002).
[4] N. M. R. Peres, F. Guinea and A. H. Castro Neto, Phys. Rev. B **73**, 125411 (2006).
[5] V. P. Gusynin and S. G. Sharapov, Phys. Rev. Lett. **95**, 146801 (2005).
[6] M. I. Katsnelson, Eur. Phys. J. B **51**, 157 (2006).
[7] K. Nomura and A. H. MacDonald, report cond-mat/0606589.
[8] I. L. Aleiner and K. B. Efetov, report cond-mat/0607200.
[9] V. V. Cheianov and V. I. Fal'ko, Phys. Rev. B **74**, 041403 (2006).
[10] C. W. J. Beenakker, Phys. Rev. Lett. **97**, 067007 (2006).
[11] H. B. Heersche, P. Jarillo-Herrero, J. B. Oostinga, L. M. K. Vandersypen and A. F. Morpurgo, Nature **446**, 56 (2007).
[12] A. Shailos, W. Nativel, A. Kasumov, C. Collet, M. Ferrier, S. Guéron, R. Deblock and H. Bouchiat, report cond-mat/0612058.
[13] A. Yu. Kasumov, R. Deblock, M. Kociak, B. Reulet, H. Bouchiat, I. I. Khodos, Yu. B. Gorbatov, V. T. Volkov, C. Journet and M. Burghard, Science **284**, 1508 (1999).
[14] M. Titov and C. W. J. Beenakker, Phys. Rev. B **74**, 041401(R) (2006).
[15] J. González, F. Guinea and M. A. H. Vozmediano, Nucl. Phys. B **424**, 595 (1994).
[16] J. González, F. Guinea and M. A. H. Vozmediano, Phys. Rev. B **59**, R2474 (1999).
[17] It can be easily seen that the relative conductance Nt^2W/v_F is a dimensionless quantity, as the electron fields $\Psi^{(a)}$ in graphene have the dimension of the inverse of a length according to Eq. (1).
[18] J. González, Phys. Rev. Lett. **87**, 136401 (2001).
[19] J. González, J. Phys.: Condens. Matter **15**, S2473 (2003).
[20] See, for instance, S. A. Chin, Ann. Physics **108**, 301 (1977).

[21] A. A. Abrikosov, L. P. Gorkov and I. E. Dzyaloshinski, *Methods of Quantum Field Theory in Statistical Physics*, Chap. 7 (Dover, New York, 1975).
[22] P. Dubos, H. Courtois, B. Pannetier, F. K. Wilhelm, A. D. Zaikin and G. Schön, Phys. Rev. B **63**, 064502 (2001).
[23] T. Ando, J. Phys. Soc. Jpn. **74**, 777 (2005).